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# PATENT COOPERATION TREATY

# PCT

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

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|---|---|---|
| Applicant's or agent's file reference<br>FP17555 | **FOR FURTHER ACTION** | See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416). |
| International Application No.<br>**PCT/AU2003/000385** | International Filing Date *(day/month/year)*<br>28 March 2003 | Priority Date *(day/month/year)*<br>28 March 2002 |
| International Patent Classification (IPC) or national classification and IPC<br>**Int. Cl.** [7] C08K 3/34 | | |
| Applicant<br>COMPCO PTY LTD et al | | |

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 3 sheets, including this cover sheet.

   [X] This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of **18 sheet(s)**.

3. This report contains indications relating to the following items:

   I [X] Basis of the report
   II [ ] Priority
   III [ ] Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   IV [ ] Lack of unity of invention
   V [X] Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   VI [ ] Certain documents cited
   VII [ ] Certain defects in the international application
   VIII [ ] Certain observations on the international application

| | |
|---|---|
| Date of submission of the demand<br>10 October 2003 | Date of completion of the report<br>11 March 2004 |
| Name and mailing address of the IPEA/AU<br>AUSTRALIAN PATENT OFFICE<br>PO BOX 200, WODEN ACT 2606, AUSTRALIA<br>E-mail address: pct@ipaustralia.gov.au<br>Facsimile No. (02) 6285 3929 | Authorized Officer<br>A Yong<br>**ALBERT S. J. YONG**<br>Telephone No. (02) 6283 2160 |

**I. Basis of the report**

1. With regard to the **elements** of the international application:*

☐ the international application as originally filed.

☒ the description, pages **1-3,5,7,12,14-19,22,24,26-27,29-32,44 (abstract)** as originally filed,
pages , filed with the demand,
pages 4,6,8-11a,13,20-21,25,28 received on **30 December 2003** with the letter of **29 December 2003**
page 23 received on **9 March 2004** with the letter of **9 January 2004**

☒ the claims, pages **33,35,37-40** , as originally filed,
pages , as amended (together with any statement) under Article 19,
pages , filed with the demand,
pages 34, 43 , received on **30 December 2003** with the letter of **29 December 2003**
pages 36,41-42 received on 9 March 2004 with the letter of 9 January 2004

☐ the drawings, pages , as originally filed,
pages , filed with the demand,
pages , received on with the letter of

☐ the sequence listing part of the description:
pages , as originally filed
pages , filed with the demand
pages , received on with the letter of

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.
These elements were available or furnished to this Authority in the following language which is:

☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).

☐ the language of publication of the international application (under Rule 48.3(b)).

☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

4. ☐ The amendments have resulted in the cancellation of:

☐ the description, pages
☐ the claims, Nos.
☐ the drawings, sheets/fig.

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* *Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).*

** *Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report*

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

| | | |
|---|---|---|
| Novelty (N) | Claims 1-83 | YES |
| | Claims | NO |
| Inventive step (IS) | Claims 1-83 | YES |
| | Claims | NO |
| Industrial applicability (IA) | Claims 1-83 | YES |
| | Claims | NO |

2. Citations and explanations (Rule 70.7)

**CITATIONS**

D1. WO 2001/096467A

D2. WO 2001/087596A

D3. WO 2001/087566A

D4. US 2001/0033924A

D5. WO 2001/066627A

D6. WO 2001/010948A

D7. WO 1999/047598A

D8. US 5952093A

D9. US 5910523A

**NOVELTY AND INVENTIVE STEP**

Claims 1-83: The claimed invention relates to a composition comprising a cross-linkable and/or cross-linked ethylene (co)polymer and an intercalated nanofiller.

None of the above citations individually, or obvious combinations thereof, teaches the claimed invention. The closest art, WO 2001/096467, discloses a composite material comprising propylene graft copolymers and smectite clay. The use of cross-linked polymers in the compositions of the present invention improves the mechanical and chemical properties of the resulting products. Hence, the claims are novel and inventive in the light of the above citations.

intercalated nanofiller with a cross-linkable and/or grafted ethylene (co)polymer.

In another aspect of the process, the ethylene (co)polymer and/or nanofiller are subjected to grafting either before, during or after the mixing and delaminating and/or exfoliating step(s). The grafting preferably involves treating the ethylene (co)polymer and/or nanofiller with an organic silane which is then grafted onto the (co)polymer and/or intercalated into the nanofiller using a free radical initiator.

The present invention further provides an article which is wholly or partly composed of the nanofiller composition defined above.

In a further aspect, the present invention provides a process for preparing the article defined above which comprises either:

(a) forming or shaping the nanofiller composition defined above;

(b) combining at least one layer of the nanofiller composition with at least one other layer;

(c) cross-linking the nanofiller composition defined above; or

(d) heating and stretching the nanofiller composition defined above and cooling the stretched composition.

DETAILED DESCRIPTION OF THE INVENTION

Suitable ethylene (co)polymers include polyethylene and ethylene based alkene or alphaolefin copolymers, for example, high density polyethylene (HDPE), medium density polyethylene (MDPE), linear low density polyethylene (LLDPE), low density polyethylene (LDPE), very low density polyethylene (VLDPE), and ultra low density polyethylene (ULDPE); ethylene hexene copolymers and ethylene octene copolymers; butylene (co)polymers such as polybutylene and polyisobutylene; ethylene-propylene copolymers (EPM); ethylene-propylene-diene terpolymers

most preferably about 75 to about 99.9% by weight. Unless stated otherwise, it will be understood that the term "% by weight" as used herein is based on the total weight of (co)polymer.

The vinyl acetate content of the ethylene-vinyl acetate copolymer (EVA) is preferably about 3 to about 80% by weight, more preferably about 9 to about 70% by weight. The vinyl acetate content is preferably about 9 to about 30% by weight for plastomeric EVA and about 38 to about 50% by weight for elastomeric EVA.

The ethylene (co)polymer may be an elastomer or a plastomer. Plastomers and elastomers can be characterised by means of specific gravity (S.G.)or density, for example, in the case of ethylene-alpha-olefin copolymers and other properties such as the differential scanning calorimetry (DSC) melting peak, Shore A hardness and elasticity modulus. Such properties will vary depending on the type of ethylene (co)polymer and its method of manufacture and the amount of (co)monomer present. By way of example, EVA with up to about 28% VA is considered a plastomer and with above about 38% being considered an elastomer. However, generally plastomers are plastomeric and elastomers are elastomeric or thermoplastic elastomeric and flexible.

Preferably, for plastomeric cross-linkable compositions, at least about 40% to about 50% by weight, more preferably at least about 60% by weight is a plastomer with the balance being an elastomer. Examples of plastomers include polyethylene such as HDPE, MDPE, LDPE, LLDPE or VLDPE; EVA with up to about 30% vinyl acetate; EPM with up to about 25% propylene; and ethylene octane copolymers with S.G. up to about 0.886. The elastomers include ethylene octane copolymers with an S.G. of at least about 0.887; ULDPE; ethylene propylene copolymers such as terpolymers with propylene co-monomers of greater than about 30%; ethylene vinyl acetate copolymers with greater than about 38% vinyl acetate;

about 30% of the masterbatch/concentrate.

The term "intercalated" or "intercalation" is used herein in its broadest sense and refers to a platelet-like or layered structure. The layers of the nanofiller which are generally composed of silicate are treated chemically by removing some cations from between the layers and intercalated with ionic or polar substances including quaternary ammonium salts, including quaternary ammonium salts, such as, optionally substituted long chain hydrocarbon quaternary ammonium salts, for example, benzyl or alkyl substituted long chain hydrocarbon quaternary ammonium salts, alkyl substituted tallow or hydrogenated tallow quaternary ammonium salts; or bis-hydroxyethyl quaternary ammonium salts. Suitable counter anions for the quaternary ammonium cations include halides such as chloride or methyl sulphate.

The intercalated nanofiller may be an intercalated mineral nanofiller or clay which is either synthetic or natural, such as, montmorillonite, bentonite, smectite and phyllosilicate which can be or have been intercalated by organic modification with an organic intercalatent selected from the ionic or polar compounds described above and may be sold under the trade names Cloisite (Southern Clay Products), Nanofil (Sudchemie), Tixogel(Sudchemie) and Kunipia.

The organic intercalatent may be present in an amount up to about 40% by weight of the nanofiller. The weights in the description and examples refer to the nanofiller as supplied including the organic intercalatent.

It should be noted that in some instances the word "intercalation" includes the situation when intended to refer to nanofillers which have been intercalated with the organic intercalatent and the distance between their platelets is increased by a few nanometres are mixed with (co)polymer(s) and the (co)polymer molecules enter between the nano platelet layers thus further intercalating them

so that they are delaminated in the mixing process. This type of further intercalation is herein referred to as "delamination" and/or "further intercalation" / delamination / exfoliation. The step of delamination and exfoliation is very important. The effects of this step can be seen in the changes and improvements in the mechanical and thermo-mechanical and chemical and optical and X-ray diffraction properties of the compositions.

Nanofillers such as montmorillonite have an anisotropic, plate like, high aspect-ratio morphology which leads to a long and tortuous diffusion path through the structure and an improved barrier to permeation, particularly when used in combination with the cross-linked ethylene (co)polymers of the present invention.

The amount of nanofiller is about 0.1 to about 15%, preferably about 1 to about 10%, more preferably about 2 to about 6% by weight.

It will be appreciated that known fillers may optionally and/or additionally be included in the composition. Suitable known fillers include inorganic and/or mineral fillers such as clays which may be calcined; talc; mica; kaolin; alkaline earth metal carbonates, for example, calcium carbonate, magnesium calcium carbonate or hydrated basic magnesium carbonate; and metal hydroxides, for example, aluminum or magnesium hydroxide. The fillers may optionally be coated with, for example, stearic acid, stearates such as calcium stearate, silanes such as vinyl silane, siloxanes and/or organo-titanates. While such coatings can be used to coat the fillers, they can also be added simultaneously, sequentially and/or separately with the fillers.

The composition of the present invention may be subjected to (i) silane grafting; (ii) the addition of cross-linking agents; and/or (iii) radiation cross-linking at any step of the process.

(i) The silane grafting may be performed using an organic silane and a free radical initiator. In

an embodiment preferred for economical reasons, effective amounts of organic silane and peroxide are added to the (co)polymer and/or nanofiller either before or during the mixing step and then grafted onto the (co)polymer at temperatures preferably of about 160 to about 240°C, more preferably about 180 to about 230°C, most preferably about 190 to about 220°C. This grafting is carried out either in the first mixing step or in a subsequent or even in a separate mixing step, after the (co)polymer and nanofiller have been mixed. In a particularly preferred embodiment, the silane and the peroxide are added to both the (co)polymer and/or nanofiller which facilitates exfoliation and/or delamination of the nanofiller and grafted to the polymer in one step. In an alternative embodiment, the (co)polymer is grafted using the organic silane and peroxide and then mixed with the nanofiller followed by exfoliation and/or delamination.

In another embodiment, the (co)polymer(s), of which at least one has polar group(s), is or are mixed with the nanofiller for the purpose of polymer intercalation and/or delamination or exfoliation at temperatures up to about 200°C. The resulting intercalated polymer is then mixed in a second step with further (co)polymer, a free radical initiator peroxide and an organic silane and grafted onto the (co)polymer(s) at higher temperatures, preferably about 190 to about 220°C. The masterbatch of nanofiller in a (co)polymer(s) can be made with about 15 to about 45% nanofiller content. It is then subsequently mixed in a second step with further (co)polymer(s) and then grafted with peroxide and vinyl silane in the same second step or in a third step.

Suitable organic silanes include vinyl silanes, for example, vinyl alkoxy silane such as vinyl-tris-methoxy-silane (VTMOS), vinyl-tris-methoxy-ethoxy-silane (VTMOEOS), vinyl-tris-ethoxy-silane, vinyl-methyl-dimethoxy-silane and gama-methacryl-oxypropyl-tris-methoxy-silane; or long aliphatic hydrocarbon chain

silanes.

Vinyl silanes are preferred and may be added in an amount from about 0.5 to about 2.2% by weight of the (co)polymer, preferably about 0.8 to about 2%, more preferably about 1 to about 1.8% by weight.

The term "free radical initiator" is used herein in its broadest sense and refers to an unstable molecule or compound which generates free radicals. Examples of suitable initiators include peroxides such as dicumyl peroxide, di-tertiary-butyl peroxide, tertiary-butyl-cumyl peroxide and bis-tertiary-butyl-cumyl peroxide i.e., di(tert-butyl-peroxy-diisopropyl benzene) and 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane. The free radical initiator is preferably added in an amount of about 0.05 to about 0.3% by weight calculated on the amount of (co)polymer, more preferably about 0.15 to about 0.2% by weight. For this type of subsequent cross- linking the presence of moisture, water or steam is required, preferably with a catalyst added. A wider, more flexible range of ratios of peroxide to vinylsilane to be grafted is possible. The peroxide addition is possible up to about 0.5%.

Silane cross-linking is also called moisture cross-linking. After forming the article made by extrusion and/or moulding, film forming is carried out in the presence of water, steam or moisture at ambient or preferably at higher temperatures of up to about 90°C to about 100°C or higher if pressure is applied. Catalyst e.g. di-butyl-tin dilaurate (DBTDL), di-octyl-tin dilaurate (DOTDL), is added to the cross-linkable composition prior to or during forming, or it can be added to the water used for cross-linking in it as a medium.

The speed and the duration of the cross-linking will depend on the type of (co)polymer and nanofiller used in the composition, of the temperature, of the humidity or water present and of the thickness of the composition.

(ii) The (co)polymers, compositions and/or

Examples of such co-agents include unsaturated allylic compounds, triallylcyanurate, acrylic compounds and acrylate or polyacrylate compounds. Protection against radiation damage to the (co)polymer can also be achieved by the addition preferably of up to about 2% by weight of radiation protectors such as trimethyl quinoline polymers or oligomers, for example, Age Rite Resin D and Anox HB. The (co)polymer and/or composition may also be cross-linked after grafting the (co)polymer or composition with an organic silane with the aid of a free radical initiator. Catalysts for cross-linking include DBTDL (di-butyl-tin-dilaurate) or dioctyl-tin-dilaurate (DOTDL) or other known catalysts.

Radiation cross-linking may be carried out at room temperatures or rising above ambient due to the high energy radiation.

It will be appreciated that one or more additives known in the art of polymer processing can also be included in the composition and added at any stage of the process. They can be added during the mixing steps or at the stage of forming in the form of masterbatches/concentrates incorporated separately or in the catalyst masterbatch. Suitable additives include antioxidants, for example, phenolic antioxidants such as SANTONOX R marketed by Monsanto and IRGANOX 1010 which is pentaerythritol tetrakis (3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate or IRGANOX 1035 which is octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionate, Irganox B900, or process stabilisers such as Irgafox 168 marketed by Ciba-Geigy or aminic antioxidants such as Vulcanox HS and Flectol H which are polymerised 2,2,4-trimethyl-1,2-dihydroquinoline; metal deactivators and/or copper inhibitors, for example, hydrazides such as oxalic acid benzoyl hydrazide (OABH) or Irganox 1024 which is 2,3-bis-((3-(3,5-di-tert-butyl-4-hydroxyphenyl) proponyl))propiono hydrazide; UV absorbers, for example Tinuvin or HALS type UV absorbers; foaming or blowing

110°C for 1 to 2 up to 4 hours, depending of the thickness of the sample.

The testing was performed to Australian Standards (AS) which are in general harmonised with International Standards such as IEC, BS, DIN/VDE, EN (European Norms) and to ASTM test methods.

Mechanical properties were tested to above standards.

Oil resistance (O.R.) was tested to ASTM using ASTM oil nr.2, a criterion is the retention of 70% of original properties.

Environmental stress crack resistance (ESCR) was also tested to ASTM (AS)in tensioactive liquid at 50ºC, with unnotched samples. In general, results of over 100 hours are aimed to be achieved. In the case of nanocomposites and in particular cross-linked nanocomposites, results of thousands of hours e.g. 8000 hours were achieved and are still ongoing.

Hot Set test (HST) was made to AS: non-cross-linked materials, including nanocomposites would fail the test at above their melting temperatures and break anyway after short time anyway at 200°C. The requirement for cables is a maximum elongation under load of 175%. After 20 minutes and taking the load away the samples must revert to a maximal residual elongation of 15% or 25% for rubber/ elastomers.

For other applications the requirements are not so restricted.

The elongations under load could be higher.

The cross-linked or cross-linkable compositions of the present invention pass the HST.

Gel content was performed in boiling xylene to ASTM. The gel content shows that a composition has some degree of cross-linking. The main test for cross-linking is the HST. The gel content in silane grafted cross-linked materials is less related to the HST.

Impact resistance is tested to ASTM D-256 Izod

pendulum impact resistance of notched plastics.

The components used in the examples are from:

Qenos, Melbourne, Australia: for HDPE GM7655, GA7260H, HD1090, HD6025, LLDPE Alkatuff 425;

BASF Ludwigshafen, Germany: for HDPE HMW Lupolen 4261A;

Sabic, LLDPE Ladene MG200024;

DuPont, USA: EVA Elvax 470, Elvax 750, Elvax 760, MAH-HDPE Fusabond MB100D;

Sud-Chemie, Moosburg, Germany: Nanofil 15, Tixogel MP100;

SCP Southern Clay Products, Gonzales, TX, USA: Cloisite 15A, Cloisite 20A;

Crompton, USA/Switzerland: Silox VS 911, Silox VS294, Peroxide and Silane mix.

Other suppliers of similar materials e.g. Degussa, Germany, etc. CIBA, Switzerland: Antioxidants, Stabilisers: Irgafos FF168, Irganox B900;

Great Lakes Chemicals, USA: Antioxidants: Anox 20.

Compco Pty Ltd, Melbourne, Australia: Compylene Master-batches of Antioxidant: EL900140AO, Processing aid: FL90016PA5.

The proportions of the components used in the compositions of the Examples are given in % by weight of the total composition. These %s have been rounded to the first decimal point.

Comparative Example 1

HDPE Qenos GM7655 MFI 0.2, granules..................83.5%
HDPE Qenos GA7260H MFI 25, powder....................14.7%
Silox VS 911 Crompton.................................1.2%
Stabiliser Irganox 168 FF.............................0.2%
Anox 20 Great Lakes...................................0.4%

Hot Set Test (at 200°C):

Hot Set Test (at 200°C):
under load....................................100%
without load..................................0%

The addition of Tixogel has significantly improved the cross-linking of the composition compared to Example 2.

Example 3

LLDPE Alkatuff 425 granules..........................79.3%
LLDPE Ladene powder..................................14.0%
MAH-HDPE Fusabond MB 100 D...........................1.0%
Irganox 168 FF....................................... 0.2%
Anox 20..............................................0.4%
Tixogel MP 100.......................................5.1%

Flexural modulus, Mpa................................557
Tensile Strength (TS),at yield Mpa...................24.2
Tensile strength at break Mpa........................13.2
Impact resistance Izod, J/m..........................273

This composition is cross-linkable and not grafted nor cross-linked.

Example 4

HDPE HMW Lupolen 4261A powder........................88.1%
Stabiliser mix:
Irgafos 168FF+Anox20+Ca-Stearate(0.2+0.2+0.5%)........0.9%
MAH-HDPE Fusabond MB 100D.............................1.0%
Tixogel/Lupolen mix (30% Tixogel: 70% Lupolen):
Lupolen 4261A.........................................7.0%
Tixogel MP100 (via side feeder).......................3.0%

Flexural modulus, Mpa.................................531
TS at yield Mpa.......................................22.1
TS at break Mpa.......................................17.3

Composite mix**(incl. Tixogel filler) [10%]

(above 10% comp. mix** consisting of:

Tixogel MP 100(via side feeder)....................3.0%
Irgafos 168FF(process stabiliser)..................0.2%
Calcium stearate...................................0.5%
HDPE, MFI 20, MG 20224 powder......................0.9%
HDPE Lupolen 4261A.................................2.1%
Composition of Example 5...........................3.1%
Anox 20............................................0.2%
Subtotal of composite mix**.......................10.0%
Total composition example 6........................100%

*These components were pre-mixed and grafted separately.

Hot Set Test (at 200°C):

elongation under load..............................173%
elongation with load removed (relaxed)...............7%

## Example 7

HDPE MFI 10, HD1090 granules......................88.3%
LLDPE Ladene MG200024 powder.......................9.7%
Silox VS 911.......................................1.6%
Irgafos 168 FF (process stabiliser)................0.2%
Anox 20............................................0.2%

After grafting and prior to forming, 5% catalyst masterbatch/concentrate was added.

Hot Set Test (at 200°C)

Elongation under load..............................250%
Residual elongation relaxed.........................16%

Flexural modulus, Mpa..............................552
TS at yield Mpa...................................24.2
TS at break Mpa...................................15.5

Flexural modulus, Mpa............................757 851

Impact resistance Izod, J/,m.....................161 193

This composition may be grafted and with vinylsilane and peroxide for subsequent cross-linking. Alternatively, the composition may be cross-linked after peroxide addition or other cross-linking.

Example 12

EVA Elvax 760 (9.3%VA, MFI2= 2)........................83.2

LLDPE Ladene MG200024 (20MFI) powder....................10

Nanofil 15..............................................5%

Silox VS 924............................................1.8%

HST:....................................................50%

Example 13

MAH-HDPE Fusabond MB100D................................83.2%

LLDPE Ladene MG200024 (MFI20)...........................10%

Nanofil 15..............................................5%

Silox VS 924............................................1.8%

TS at yield, Mpa........................................25.2

TS at break, Mpa........................................15.0

Flexural modulus, Mpa...................................654

O.R.(oil resistance), TS @ break retained:..............99%

O.R. EB (elongation @ break) retained...................95%

Gel content:............................................28.8%

Impact resistance Izod, J/m.............................103

Example 14

MAH-HDPE Fusabond MB100D................................83.2%

LLDPE Ladene MG200024 (MFI20)...........................10%

Nanofil 15 (dried)......................................5%

Silox VS 924............................................1.8%

TS at yield, Mpa........................................25.0

grafted with compounds containing carboxylic acid or anhydride group(s).

7. A composition according to claim 6, in which the carboxylic acid or anhydride group is maleic anhydride or acid or fumaric anhydride or acid.

8. A composition according to claim 6 or claim 7, in which the grafted ethylene (co)polymer is a maleic anhydride (MAH) or maleic acid grafted copolymer.

9. A composition according to claim 8, in which the maleic anhydride (MAH) or maleic acid grafted copolymer is LDPE-MAH, HDPE-MAH, EP-MAH, EPR-MAH, PE-MAH or PP-MAH.

10. A composition according to any one of claims 1 to 9, in which the ethylene (co)polymer contains polar group(s).

11. A composition according to claim 10, in which the polar group(s) are carboxylic group(s), maleic group(s) and/or ester group(s).

12. A composition according to claim 10 or claim 11, in which the amount of (co)polymer with polar group(s) is about 0.01% of the total (co)polymer.

13. A composition according to any one of claims 10 to 12, in which the amount of (co)polymer with polar group(s) is about 0.5% of the total (co)polymer.

14. A composition according to any one of claims 10 to 13, in which the amount of (co)polymer with polar group(s) is at least about 5% of the total (co)polymer.

15. A composition according to any one of claims 10 to 14, in which the amount of (co)polymer with polar

VLDPE, EVA with up to about 30% vinyl acetate, EPM with up to about 25% propylene and/or an ethylene octane copolymer with S.G. up to about 0.886.

25. A composition according to any one of claims 21 to 24, in which the elastomer is an ethylene octane copolymer with an S.G. of at least about 0.887, ULDPE, ethylene propylene copolymer, an ethylene vinyl acetate copolymer with greater than about 38% vinyl acetate, EPDM, EPM, and/or EPR.

26. A composition according to claim 25, in which the ethylene propylene copolymer is a terpolymer with a propylene co-monomer of greater than about 30%.

27. A composition according to claim 35, in which the vinyl acetate content for plastomeric EVA is about 9 to about 30% by weight.

28. A composition according to claim 25, in which the vinyl acetate content for elastomeric EVA is about 38 to about 50% by weight.

29. A composition according to any one of the preceding claims, in which the cross-linkable and/or cross-linked ethylene (co)polymer forms at least about 40% by weight of the total weight of (co)polymer.

30. A composition according to any one of the preceding claims, in which the nanofiller has particle(s) in the order of size of less than 50nm.

31. A composition according to any one of the preceding claims, in which the thickness of the nanofiller particles is about 1nm to about 100nm.

32. A composition according to any one of the

preceding claims, which further comprises one or more additives known in the art of polymer processing.

68. A composition according to claim 67, in which the additive is an antioxidant, metal deactivator, copper inhibitor, UV absorber, foaming or blowing agent which is either endothermic or exothermic, processing and/or thermal stabiliser, pigment, flame retardant, extender, plasticiser and/or softener.

69. A process for preparing a cross-linkable and/or cross-linked nanofiller composition which comprises either:

(a) mixing and delaminating and/or exfoliating in one step a cross-linkable and/or cross-linked ethylene (co)polymer and an intercalated nanofiller;

(b) mixing a cross-linkable ethylene (co)polymer with an intercalated nanofiller; and

delaminating and/or exfoliating at least part of the nanofiller; or

(c) delaminating and/or exfoliating at least part of an intercalated nanofiller; and

mixing the delaminated and/or exfoliated intercalated nanofiller with a cross-linkable and/or cross-linked ethylene (co)polymer.

70. A process according to claim 69, in which the ethylene (co)polymer and/or nanofiller are subjected to grafting either before, during or after the mixing and/or exfoliating and/or delaminating step(s).

71. A process according to claim 70, in which the grafting involves treating the ethylene (co)polymer and/or nanofiller with an organic silane which is then grafted onto the (co)polymer and/or intercalated into the nanofiller.

72. A process according to claim 71, in which the organic silane is grafted using a free radical initiator.

73. A process according to any one of claims 69 to 72, which further comprises the step of cross-linking the (co)polymer before step (a) or cross-linking the composition after step (b) or (c).

74. A process according to claim 73, in which the composition and/or ethylene (co)polymer is silane cross-linked, cross-linked by adding a cross-linking catalyst, silane cross-linked or radiation cross-linked.

75. A process according to any one of claims 69 to 74, in which the (co)polymer is granulated, pelletised, powderised, cut and/or diced.

76. A process according to any one of claims 69 to 75, in which the (co)polymer and the nanofiller are pre-mixed or added simultaneously, sequentially and/or separately to a mixing apparatus.

77. A process according to any one of claims 69 to 76, in which the nanofiller or composition are exfoliated and/or delaminated using high shear processing.

78. A process according to any one of claims 69 to 77, in which a further exfoliation and/or delamination step is performed at any stage of the process.

79. A process according to any one of claims 69 to 78, in which other fillers and/or additives are added simultaneously, sequentially and/or separately at any step of the process.

80. A process according to claim 79, in which the (co)polymer, nanofiller, other fillers and/or additives

are dry or dried in a separate step prior to step (a).

81. An article which is wholly or partly composed of the nanofiller composition defined in any one of claims 1 to 68.

82. An article according to claim 81, which is a profile, tube, pipe, film, sheet, tile, floor covering, container or packaging for food.

83. A process for preparing the article defined in claim 81 or claim 82, which comprises either:

(a) forming or shaping the nanofiller composition defined in any one of claims 1 to 68; or

(b) combining at least one layer of the nanofiller composition defined in any one of claims 1 to 68 with at least one other layer;

(c) cross-linking the nanofiller composition defined in any one of claims 1 to 68; or

(d) heating and stretching the nanofiller composition defined in any one of claims 1 to 68 and cooling the stretched composition.